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ANTIMONY OXIDE MINERALS FROM HUNGARY

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ABSTRACT

Most of the Hungarian stibnite and fahlore occurrences contain antimony-bearing weathering products that earlier were either unidentified or their identification was doubtful. All these occurrences were investigated by using XRPD and EDS for the identification of the substances in question.

The results of investigations are reported here together with a brief description of the Hungarian stibnite occurrences, the antimony-bearing weathering products and their mineral assemblages. Occurrences of stibnite and antimony oxides are shown in *Fig. 1.* A list of Hungarian stibnite localities arranged according to mountains is given in Table 1 and a list of antimony oxide minerals is given in Table 2

Cervantite, in spite of being reported most frequently from this mineral group in the Hungarian mineralogical literature, has been found to occur only in Gyöngyösoroszi. On the other hand, stibiconite proved to be ubiquitous, and several new occurrences of bindheimite were also found. Our investigations confirmed the occurrences of valentinite and tripuhyite in Hungary and the first occurrence of partzite is also described.

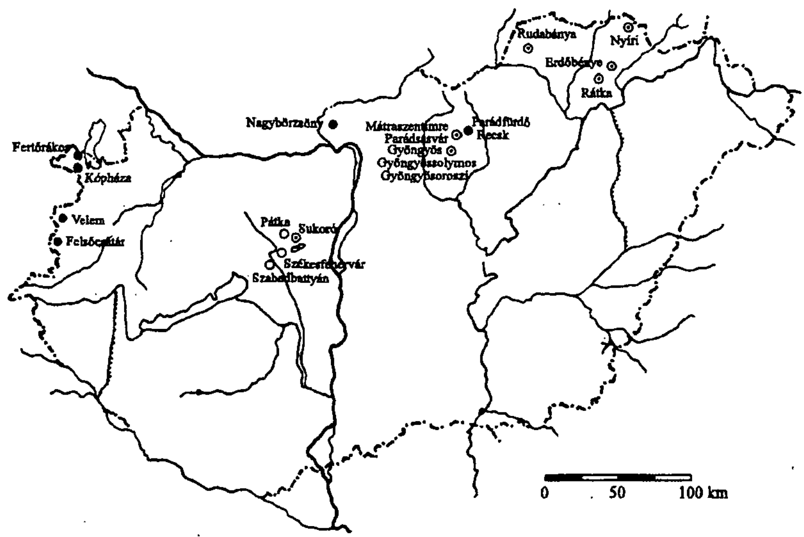

Fig. 1. Occurrences of stibnite and "antimony ochres" in Hungary
Symbols: ● stibnite, ⊙ stibnite and antimony ochre, O antimony ochre

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TABLE 1

Stibnite occurrences of Hungary (arranged by mountains from the west)

	LOCALITY	AREA	REFERENCES (other than this paper)
2.1.1	* Felsőcsatár: talc mine	Kőszeg–Vashegy	HOM collection
2.1.2	? Velem: Szent Vid Hill	Kőszeg–Vashegy	Földvári et al. (1949); Bendefy (1963, 1968); Vendel (1969)
2.1.3	* Velem: V-3, 4, 5, 6 boreholes	Kőszeg–Vashegy	Nagy (1972)
2.2.1	* Kópháza: Kő Hill	Sopron Hills	HOM collection
2.2.2	* Fertőrákos: Gödölye Hill	Sopron Hills	Fazekas et al. (1972)
2.3.2	* Székesfehérvár II inclined borehole	Velence Hills	Mikó (1964)
2.3.3	Pátka: Kőrakás Hill	Velence Hills	Jantsky (1952); Kiss (1954); Kaszanitzky (1959)
2.3.5	Lovasberény: Meleg Hill	Velence Hills	Jantsky (1952, 1957); Kubovics (1958)
2.4.1b	* Nagybörzsöny: Nb-13 borehole	Börzsöny Mts.	Nagy (1984)
2.5.1	* Recsk: –900 m level	Mátra Mts.	HOM collection
2.5.2	* Parádfürdő: Fehér-kő Hill / Hegyes Hill	Mátra Mts.	HOM collection / Nagy (1985)
2.5.3a	Gyöngyösoroszi: Aranybánya-bérc, Arany-Péter, Bányabérc, Hidegkút, Károly, Kiskút, Új-Károly veins	Mátra Mts.	Vidacs (1961a); Koch (1966); Kun (1985); Dódony (1986); Nagy (1986)
2.5.3b	Mátraszentimre: Mátraszentimre vein / outcrops on the Teréz Hill	Mátra Mts.	Koch (1966); Nagy and Barbácsi (1966); Csongrádi (1969); Kun (1985) / MÁFI Collection
2.5.4	Gyöngyössolymos: Asztag Kő Hill	Mátra Mts.	Szurovy (1940); Kiss (1960); Koch (1966); Szakáll (1989)
2.5.5	Parádsasvár: road cut	Mátra Mts.	Nagy and Szentes (1969)
2.5.6	Gyöngyös–Mátraháza road cut	Mátra Mts.	HOM collection
2.6.1	Rudabánya: Polyánka and Andrássy III. mines	Rudabánya Mts.	Szakáll et al. (in press)
2.6.2.	? Martonyi	Rudabánya Mts.	Maderspach (1880)
2.7.1	Erdőbénye: Ligetmajor and Mogyorósok areas / Sás stream valley	Tokaj Mts.	Endes (1988); Szakáll (1991) / Szabó (1870); Papp (1992)
2.7.2	Rátka: Hercegköves / Koldu open pits	Tokaj Mts.	Jánosi and Papp (1985) / MÁFI collection
2.7.3a	? Telkibánya: old references	Tokaj Mts.	Zipser (1817); Cotta and Fellenberg (1862); Szakáll et al. (1994)
2.7.3b	Telkibánya: Nyíri gallery	Tokaj Mts.	HOM collection

Symbols: ? – uncertain information; * – without antimony oxides

Abbrevations: MÁFI – Hungarian Geological Institute, Budapest; HOM – Herman Ottó Museum, Miskolc

TABLE 2

Antimony oxide minerals of Hungary (arranged by mountains and localitites within)

	stibiconite	**bindheimite**	**partzite**	**tripuhyite**	**cervantite**	**sénarmontite**	**valentinite**
Localities	$SbSb_2O_6OH$	$Pb_2Sb_2O_6(O,OH)$	$CuSb_2(O,OH)_7$(?)	$FeSb_2O_6$	Sb_2O_4	Sb_2O_3	Sb_2O_3
Szabadbattyán		■					
Pátka, Kőrakás Hill	■						
Pátka, Szűzvár		■					
Lovasberény, Meleg Hill	■	■					
Parádfürdő	■	■					
Gyöngyösoroszi	■				■		
Mátraszentimre	■						
Gyöngyössolymos, Asztag-kő Hill	■			■		■	
Parádsasvár	■						
Gyöngyös-Mátraháza	■						
Rudabánya	■	■	■				■
Erdőbénye	■						
Rátka, Hercegköves	■			■			
Rátka, Koldu	■						

INTRODUCTION

Antimony oxides – or "antimony ochres" as they have usually been called – are formed in the oxidation zone of ore deposits and showings as weathering products of stibnite or occasionally of other antimony-bearing sulphides. Weathering of stibnite starts at the cleavage planes and can easily proceed turning the whole mass of the crystal into antimony oxides. Therefore antimony oxides (first of all stibiconite) frequently appear as pseudomorphs after stibnite. Bindheimite and (less commonly) stibiconite are formed by the weathering of Pb, Sb, sulphides (usually tetrahedrite or Pb, Sb, sulphosalts). They appear first in scattered grains, later in small aggregates and finally turn the sulphide into a spongy mass. Since bindheimite is usually formed during the weathering of different, tightly intergrown sulphides, it is as a rule accompanied by other secondary minerals (mainly cerussite and jarosite).

"Antimony ochres" are difficult to identify due to their fine-grained habit and frequent presence of admixed minerals. Despite this fact species names were assigned to such substances in the past without adequate research. In some cases a re-examination of the mineral in question proved that the earlier identification was false. In addition to these misinterpreted old occurrences, some new unknown ones were discovered in the last decade. This is why we undertook a comprehensive survey of antimony oxide minerals of Hungary.

X-ray powder diffraction (XRPED) and electron microprobe (SEM-EDS) was used for the identification of the minerals. Unfortunately, weathering products of stibnite frequently show a low degree of crystallinity (cf. Vitaliano and Mason, 1952). The interpretation of the XRPD pattern may be difficult even if the degree of crystallinity is sufficient, when the sample is an unseparable mixture. The strongest reflections of some antimony oxide minerals are close to each other (in addition to those belonging to the same structural group). For example the two most intensive reflections of cervantite almost coincide with two strong peaks of stibiconite. VITALIANO and MASON (1952) have already pointed out the difficulties of the detection of small quanitites of valentinite associated with stibiconite. Some frequent associated minerals (stibnite, quartz etc.) may also cause problems; an example is the detection of subordinate tripuhyte in the presence of quartz and stibiconite.

In this paper only the minerals that were detected reliably, i.e. at least three or four characteristic reflections were present on the X-ray pattern of the specimen (some 70 to 80 samples were studied by XRPD), are discussed. The XRPD studies were completed with semi-quantitative EDS analyses. Chemical data helped us to detect antimony oxides and to distinguish structurally related but chemically different phases (e. g. stibiconite, bindheimite and partzite).

EXPERIMENTAL

X-ray powder diffraction (XRPD) investigations:

Dept. of Mineralogy, Eötvös L. University (hereafter reffered to as ELTE): Siemens 500 D diffractometer, CuK_α radiation, accelerating voltage 40 kV, tube current 20 mA, graphite monochromator, scan speed 1 or 2°20/min.

X-ray laboratory, MOL Rt. Philips PW 1820 diffractometer, CuK_α radiation, accelerating voltage 40 kV, tube current 30 mA, graphite monochromator, scan speed 0,05° 20/sec.

X-ray laboratory, Bay Zoltán Institute of Materials Science and Technology: Philips PW/1820 diffractometer, CuK_α radiation, accelerating voltage 45 kV, tube current 30 mA, graphite monchromator, scan speed 0.04°20/sec.

Electron microprobe (SEM-EDS) investigations:

Dept. of Metallurgy, University of Miskolc: AMRAY 1830i scanning electron microscope, EDAX 9900 energy dispersive X-ray spectrometer, accelerating voltage 20 kV, sample current 10–10 A, SiLi detector.

SURVEY OF STIBNITE AND ANTIMONY OXIDE OCCURENCES BY AREAS

Occurrences are described by mountains approximately in a west to east order. Stibnite localities without antimony-containing weathering products are also included for the sake of completeness. For every area a brief geological outline is given first on the basis of BÉRCZI and JÁMBOR (1998), CSÁSZÁR (1998), MOLNÁR et al. (1999) and other handbooks. More detailed information about the localities themselves based mainly on the data of JANTSKY (1966) and MOLNÁR et al. (1999) is included in the relevant entries.

Kőszeg–Vashegy area

Metamorphic rocks of the small Kőszeg Hills and Vashegy areas belong to the Rechnitz and Eisenberg tectonic windows of the Eastern Alps, respectively. They are made up by Penninian formations, metasediments and metamorphosed ophiolites. Sulphide mineralisations are scarse and little known, but stibnite is present in some occurrences.

Felsőcsatár, talc mine, stibnite occurrence

In a presumably Jurassic greensicht formation serpentinite, talc schist, chlorite schist, tremolite schist etc. are found at Felsőcsatár. A mine between 1952–1995 exploited the talc deposit. Quartz veins in the schists host late galena- and sphalerite-bearing sulphide showings. A stibnite sample from this paragenesis is stored in the Herman Ottó Museum (hereafter referred to as HOM) collection (inv. # 21 871). Acicular aggregates of stibnite are associated with an unidentified Pb, Sb sulphosalt. Weathering products of stibnite have not been observed yet.

Szent Vid Hill, Velem, doubtful stibnite occurrence

BENDEFY (1963, 1968) claimed that Bronze Age inhabitants of the settlement on Szent Vid Hill had mined antimony, iron and manganese. He found stibnite-bearing quartzite pebbles on the hill and observed outcrops of stibnite-bearing quartz veins. Geologists who mapped the area (FÖLDVÁRI et al, 1948; LENGYEL, 1953; VENDEL, 1969) have never found such outcrops and, therefore, they supposed that the stibnite-bearing material had been transported from the nearby stibnite deposit at Schlaining or the actually occurring pyrolusite was confused with stibnite. The newest geoarcheological studies (CZAJLIK et al, 1995) proved that the source of ore for production of antimony-bearing bronze tools is from ancient mines close to Salzburg or from the Carpathians. Our repeated attempts to find stibnite specimens were also unsuccessful, moreover, there are no such specimens in any public collection. By this reason we think that the data on the occurrence of stibnite on the Szent Vid Hill are false.

Velem, boreholes, ore showings

Disseminated pyrite and marcasite grains were found in a presumably Jurassic greenschist-facies calc-phyllite formation (NAGY, 1972). The ore microscopic studies of cores from V–3, 4, 5, 6 boreholes by Béla Nagy revealed magnetite, chalcopyrite, native gold, stibnite, galena and sphalerite in addition to the iron sulphides. Stibnite is the latest precipitation product of the latest post-tectonic ore-forming phase. No weathering products have been reported so far.

Sopron Hills

Metamorphic rocks of Sopron Hills belong to the Lower Austroalpine "Grobgneis" complex. This polymetamorphic crystalline complex is made up by metamorphosed sedimentary and magmatic rocks. Only one occurrence is known among the rare sulphide mineralisations where stibnite was found. Metamorphic rocks of the nearby "Fertőrákos Schist Isle" (the Hungarian aprt of the "Mörbisch Schist Isle") have been formed by polymetamorphism of sedimentary and igneous rocks of varied composition ranging from acidic to basic. It is correlated with the Wechsel series. Different levels of the series contain sulphide ore mineralisations (KOCH, 1985).

Kő Hill, Kópháza, stibnite occurrence

A stibnite sample from an outcrop of the Palaeozoic Sopron gneiss at the Kő Hill near Kópháza is found in the HOM collection (inv. # 21702). Rare sulphide clusters up to 0.5–1 cm) contain acicular stibnite crystals (up to 0.1–0.5 mm) associated with marcasite, galena and sphalerite. Weathering products have not been observed yet.

Gödölye Hill, Fertőrákos, ore showings

Uraniferous schists outcrop in the vicinity of Fertőrákos near the Austrian boundary. The prospect boreholes discovered a sequence of mica schists, gneisses, amphibolites, amphibole schists etc. Different levels of the series contain peculiar polymetallic ore showings with pyrite, arsenopyrite, Co, Ni arsenides, pyrrhotite, chalcopyrite, galena, sphalerite, fahlore, U, Ti oxides etc. as main components (FAZEKAS et al., 1972; KOCH, 1985). Stibnite was found by Károly Várszegi along a veinlet in a pegmatoid body (KOCH, 1985).

Velence Hills and their surroundings

From the Szabadbattyán area, SW from the Velence Hills, Lower Carboniferous limestones are known from galleries and boreholes, having tectonic contacts with underlying Ordovician slates and overlying metamorphosed Devonian limestones. The Devonian limestones show hydrothermal metasomatic alterations and host a fahlore-containing galena deposit.

The Velence Hills are made up by Carboniferous biotite orthoclase granite that intruded an anchimetamorphic Lower Paleozoic slate series. In the eastern part the grainite body is bordered by Eocene andesite rocks. In some places the Velence granite shows considerable post-magmatic and hydrothermal alterations. Hydrothermal quartz-fluorite and quartz-barite veins with polymetallic mineralisation are frequent and in two areas near Pátka form small ore deposits with stibnite and/or fahlore as antimony minerals.

Szár Hill, Szabadbattyán, lead deposit

In the Szár Hill a metasomatic lead deposit is found in Devonian crystalline limestone along a NW–SE striking faulted zone. The limestone itself suffered a strong iron metasomatism. The most important ore mineral is galena with some fahlore, sphalerite and chalcopyrite; the main secondary minerals are cerussite, malachite, azurite and yellow, bindheimite-like ochre. A small-sclae lead mine was in operation between 1938–46 and 1949–54 (JANTSKY, 1966).

KOCH (1943) found minor amounts of a yellow, pulverulent material in close connection with cerussite. He detected Al, Fe, Pb and Si by wet tests in the mineral, which was then classified as a hydrous Al silicate with Fe and Pb as impurities. The "bindheimite-like" mineral that was described by ZSIVNY (1951) is very likely the same substance. It occurred as lemon to orange yellow, pulverulent, earthy fracture fillings in strongly cerussitised ore samples. XRPD analyses of several samples from the collections of the ELTE and the HOM justified that the yellow powdery aggregates are composed of bindheimite. Bindheimite was found not only in the cavities and fissures of the cerussitised galena but also in the accompanying limonite. It should be mentioned that due to sample preparation problems the detection of bindheimite was difficult, because most of its reflections were overlapped by those of cerussite. However, some of the samples (e.g. C97) have low cerussite content and every important reflection of bindheimite could easily be identified: 3.005 (100), 2.612 (30), 1.843 (55), 1.571 (30) [d(Å) (I_{rel})] (Fig. 2). Bindheimite is always accompanied by cerussite, other close associates are quartz and a jarosite-group mineral. XRF analyses prove a persistent iron content.

Recalling Zsivny's paper, the uncertainty of his identification of the Kőrakás Hill bindheimite was due to the difficulties in the interpretation of wet chemical data (PbO 63.42, Fe_2O_3 2.78, Sb_2O_5 24.55, H_2O 7.00, Al_2O_3 0.26, SiO_2 1.43, total 99.33%), showing excess lead and less antimony as compared to the theoretical values of bindheimite.

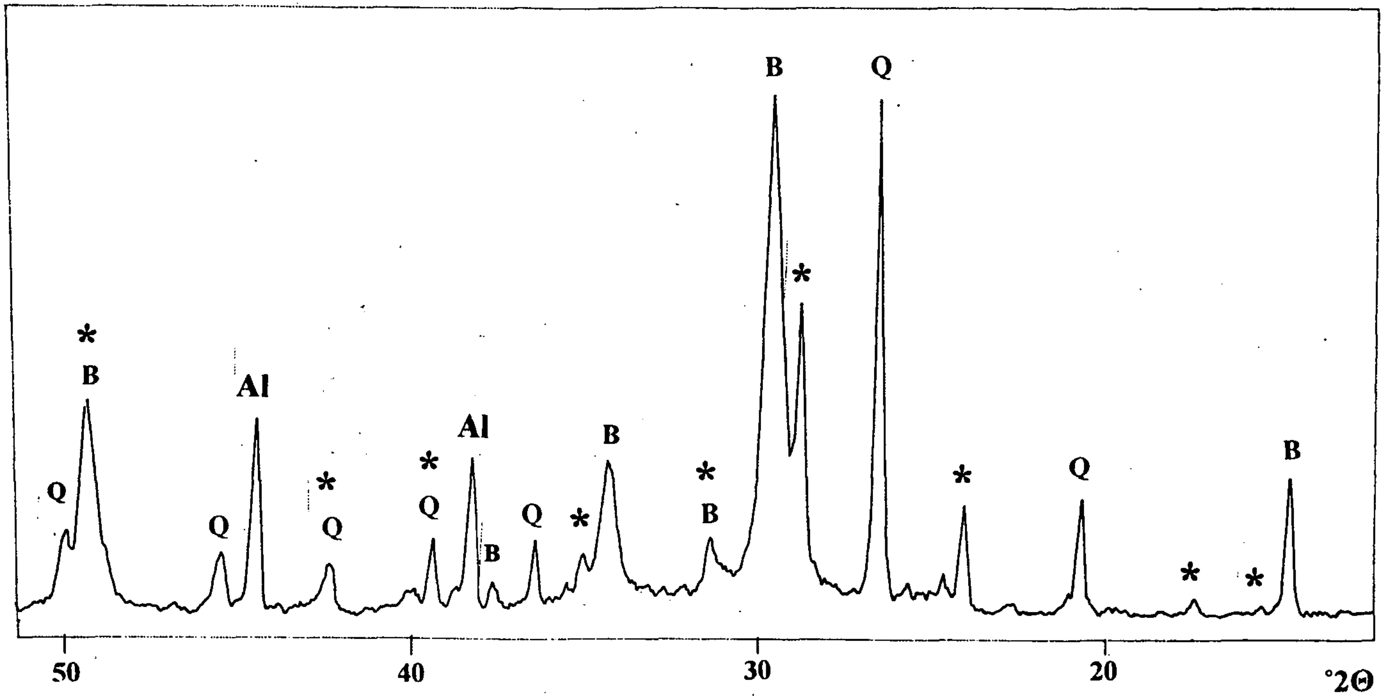

Fig. 2. XRPD pattern of bindheimite from Szár Hill, Szabadbattyán (C97)
Symbols: Al: sample holder, B: bindheimite, Q: quartz, * jarosite-group mineral

Unfortunately, the analytical details have not been published but the contradiction can be resolved if we assign some 5% of H_2O, presumably calculated from weight loss, to cerussite as CO_2. This explanation is in line with our experiences; namely we were practically unable to prepare a cerussite-free sample for XRPD analysis.

Székesfehérvár II inclined borehole

The Székesfehérvár II inclined borehole that was drilled to prospect the quartz vein of the Székesfehérvári szőlők area traversed a 2 cm thick quartz string as 199 m relative depth. The quartz string was bordered by a sphalerite I – galena – sphalerite II – stibnite – pyrite paragenesis associated with unspecified secondary minerals (MIKÓ, 1964).

Kőrakás Hill, Pátka, polymetallic ore deposit

On the Kőrakás Hill 4 km SSW from Pátka hydrothermal veins are found in silicified granite. The main components of the brecciated, banded and cockade ore are sphaleriteand galena with some chalcopyrite, stibnite and fahlore. The most important gangue minerals are quartz and fluorite; secondary minerals include cerussite, azurite, malachite, cinnabar, antimony ochre etc. (JANTSKY, 1966). The Kőrakás Hill mine operated from 1951 until 1973.

The occurrence of stibnite was first reported by JANTSKY (1952). According to KISS (1954), stibnite was found in the upper zone of the ore deposit uncovered by the first exploratory shaft (also called Földvári shaft), as radiated aggregates in vugs and fissures of the siliceous veins in granite. It was frequently replaced by antimony ochre pseudomorphs. Kiss also reported "rare powdery coatings composed of cinnabar and Pb, Sb ochre" associated with azurite and malachite (the unaltered ore was galena with microscopic sphalerite, chalcopyrite and fahlore grains). Stibnite was also found in quartz veins at the contact zone of granite and schist exposed by the north-eastern drift. According to KISS (1954) the Szűzvár stibnite forms striated aggregates composed of crystals few mm to few cm long, the crystals are rimmed or occasionally fully replaced by a yellowish weathering product. KASZANITZKY (1959) referred to this materials as cervantite although it has not been studied up to the present.

XRPD study of specimen # BE 21755 (collected by János Kiss and stored at the ELTE revealed that this substance is a well-crystallised stibiconite (sample # H145). All characteristic reflections of stibiconite appear on the record (*Fig. 3*): 5.92 (35), 3.09 (35), 2.963 (100), 2.568 (30), 1.979 (+ quartz), 1.817 (+ quartz), 1.737 (10), 1.568 (10), 1.550 (30) [d(Å) (I_{rel})]. Samples 4997, 4998 and 4999 of the MÁFI (collected by Béla Jantsky) came from the exploration workings driven on the Kőrakás Hill. These specimens contain thin stibnite bands in weathered, argillised granite. Stibnite may also form clusters of 0.5–1.5 cm in diameter along thin quartz veinlets in silicified granite. In the marginal zones of the veins and clusters, stibnite is always covered by yellowish brown, light brown weathering products, froming crusts or earthy aggregates. An XRPD pattern (sample D11) indicated the presence of stibiconite on the basis of the reflections: 5.95 (10), 3.09 (30), 2.99 (100), 2.589 (25), 1.819 (+quartz) [d(Å) (I_{rel})].

Szűzvár, Pátka, polymetallic ore deposit

The base metal vein-type mineralisation was discovered in 1951. The veins are characterised by a chalcopyrite-sphalerite-galena+tetrahedrite succession. Gangue minerals are fluorite and/or quartz with some barite. In most parts the lode is characterised by fluorite gangue, and the main products of the Szűzvár mine (operated from 1951 until

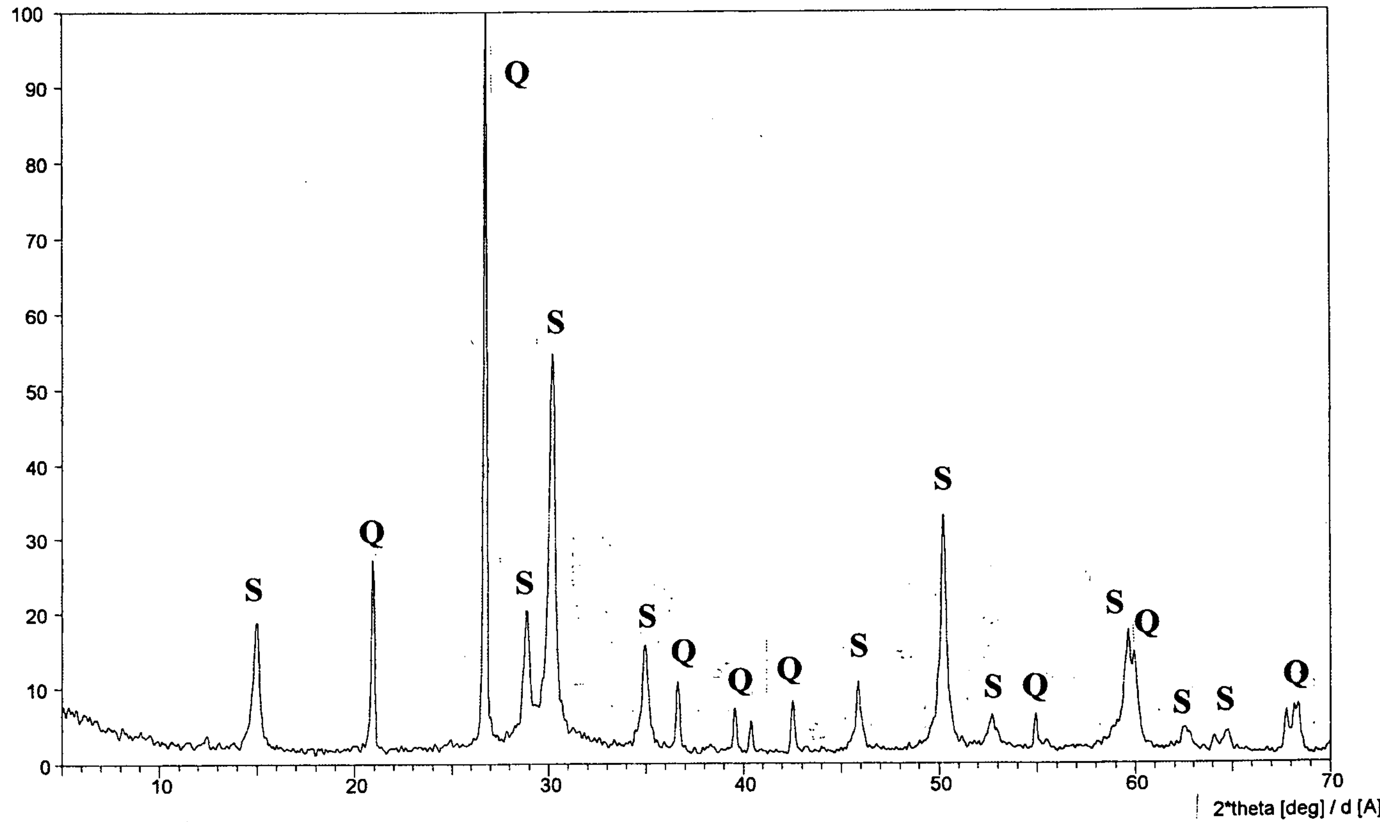

Fig. 3. XRPD pattern of stibiconite from Kőrakás Hil, Pátka (H145). Symbols: Q: quartz, S: stibiconite

1967) was in fact fluorite. Main secondary minerals are cerussite, pyromorphite, cinnabar, malachite and azurite (JANTSKY, 1966).

A "bindheimite-like" material accompanied by covellite, azurite, malachite, cerussite and cinnabar in altered fahlore-bearing galena ore was first mentioned by KISS (1954). We have also detedted this weathering product as yellow, fine-grained masses and early aggregates 1-5 mm in size on ore samples. It occurred most frequently in strongly weathered, spongy, cerussite- and pyromorphite-bearing fillings of siliceous vein outcrops above the adit. All characteristic reflections of bindheimite were identifiable on the XRPD pattern (H103) in spite of the presence of cerussite. EDS detected both antimony and lead. Associated minerals are malachite, azurite, cinnabar, linarite and cuprite.

A pale yellow pulverulent material proved to contain only antimony by EDS, the XRPD investigation of the probable antimony ochre mineral has not been completed yet.

Meleg Hill, Lovasberény, antimony showings in siliceous formations

JANTSKY (1952, 1957) found small prisms and radiating aggregates of stibnite and "yellow coatings after stibnite" together with limonite and secondary copper minerals on the SE slope of the Meleg Hill in hydrothermally silicified zones of granite. It is to be noted that some of the stibnite occurrences (viz. those on the W part of the hill) mentioned by JANTSKY (1952) are actually those of enargite. The Sb-rich zone is located as "antimonite–ochre-of-antimony area" on the map published as Fig. 1 in KUBOVICS (1958). According to ore microscopic observations of KUBOVICS (1958) stibnite forms zoned intergrowths with bismuthinite and the originally euhedral crystals are usually corroded by siliceous fluids. Stibnite is frequently replaced by unidentified antimony ochre. It is worth mentioning that we found Sb, Pb, Bi- and Pb, Bi, Ag-bearing minerals during the microprobe study of the lately collected samples. Recent studies by MOLNÁR (1995) have shown that the mineralization on the Meleg Hill is genetically connected to the Tertiary magmatic-volcanic complex bounding the area from east.

There is a stibnite-containing specimen labelled "hydroquartzite" in the collection of the Hungarian Geological Institute (hereafter referred to as MÁFI) from the eastern slope of the Meleg Hill (inv. # 4995, collected by Béla Jantsky). Columnar and needle-like stibnite crystals form aggregates 0.5 to 2 cm in size. They give a dark grey to black colour to the rock. Stibnite crystals are usually coated by a light yellow to yellowish brown weathering product, which proved to be a mixture of bindheimite and a jarosite-group mineral according the XRPD analysis (sample D47). EDS analysis (presence of K, Fe, S) suggest the presence of jarosite.

Description of recently collected samples from the eastern slope of the Meleg Hill is summarised as follows. Stibnite crystals, which are embedded in the silicified rock just as in the museum samples, reach 0.5–1 cm length and form radial-fibrous aggregates. The rim of the individual crystals is altered to white and/or light yellow antimony oxides. XRPD and EDS studies revealed the presence of stibiconite and bindheimite (samples K87–89). Associated minerals are acanthite (crusts and clusters few mm in size), jarosite (yellow, powdery aggregates), barium-pharmacosiderite (hexahedra 0.01–0.03 mm in size), segnitite and/or beudantite (dark brown crusts) and goethite.

Börzsöny Mts.

Early Middle Miocene submarine andesite extrusive domes and related volcanoclastic rocks overlain by remnants of andesite stratovolcano with andesite and diorite porphyry intrusions are exposed in the eroded central zone. Intrusion-related Cu (Mo) porphyry showings, base-metal stockworks and low-sulphidation type epithermal. Au occurrences are known. The base metal and gold rich ore veins at Nagybörzsöny were intensively mined during the Middle Ages.

a) Rózsa Hill mineralised area, Nagybörzsöny

In the central part of the Rózsa Hill area a precious metal-containing pyrrhotite–arsenopyrite ore deposit is known as stockwork impregnation in a propylitised dacite breccia pipe. The main ore minerals of the first ore-bearing phase are pyrrhotite, galena, sphalerite, pyrite and chalcopyrite, those of the second phase are arsenopyrite with native bismuth, bismuth sulphides, Pb, Bi sulphosalts and native gold. The southern part is characterised by thin polymetallic veins with the products of the first ore-bearing phase in clayey gangue. (NAGY, 1983; KOCH, 1985)

The siliceous vein fillings of the Felső Rózsa gallery in the central part of the Rózsa Hill area contain many secondary minerals. During the preliminary study a small amount of as yet unidentified antimony oxides was found by EDS in white to pale yellow pulverulent aggregates.

b) Zálog-bérc Hill, Nagybörzsöny

Stibnite grains up to 1 mm showing twin lamellae, associated with pyrite or as inclusions in the latter were found by NAGY (1984) in polished sections of the material of the Nb-13 borehole that was drilled on the Zálog-bérc Hill, SSW from the Rózsa Hill mineralised area.

Mátra Mts.

Two major volcanic units make up the Mátra Mts. The Palaeogene Unit covers 25 km^2 in the NE part of the mountains in the environs of Recsk. There an Upper Eocene calc-alkaline volcanic sequence is intercalated with sedimentary rocks. The ore complex around Recsk contains high and low sulphidation type epithermal, porphyry copper, skarn and metasomatic replacement deposits.

The Neogene Unit extends over some 350 km2. It is formed by a Miocene calc-alkaline volcanic activity. In the Western Mátra this unit has a stratovolcanic structure with caldera and subvolcanic diorite porphyry intrusions, plus several rhyolite domes and dikes. Post-caldera andesites cover highest ridges. Volcanic rocks lie on Early Miocene sediments and tuffs. The volcanic sequence was formed in three major eruption cycles during the Lower and Middle Miocene. There are several base-metal rich low-sulphidation epithermal veins in the mountains. Low-sulphidation epithermal mineralisation related to Badenian volcanic activity occurs in two parts of the caldera structure of the Western Mátra Mts. The Gyöngyösoroszi-Mátraszentimre deposit is located in the central part and the Parádsasvár-Gyöngyössolymos mineralised zone along its eastern boundary. See MOLNÁR et al., 1999 for details and further references.

Recsk, deep levels of the deposit

At Recsk (Palaeogene Unit) a near-surface enargite-luzonite deposit in the Lahóca Hill at Recsk was minedfrom the 19th century until 1980. A subvolcanic diorite intrusion, which intruded Mesozoic sedimentary rocks during the Palaeogene volcanism, hosts another, deep-seated ore deposit. The deep deposit has been discovered by boreholes in the 1960s. During the 1970–1980s it was prospected in detail by further boreholes, two shafts and drifts. In the deep-seated ore zone a typical porphyry Cu + Mo mineralisation developed in the intrusion and skarn (Cu, Zn, and Fe) mineralisation formed along the exo/endocontacts. The carbonate host rocks contain replacement Zn, Cu, Fe ore.

Stibnite was found in the –900 m level, on the drift face of the West 3 gallery in a serpentinite-brucite-calcite (aposkarn) rock (SZABOLCS TÓTH, personal communication). Acicular aggregates up to 1–2 mm are embedded in massive coarse crystalline calcite occasionally giving a grey colour to calcite. Antimony oxides have not been found.

Parádfürdő, polymetallic mineralisations

The locality is situated in the NW and SE parts of the Palaeogene Unit of the Mátra Mts. The epithermal mineralisation of this zone is spatially related to dacite domes that intruded into the andesitic stratovolcanic sequence. The dacitic host rocks (lava flows and pyroclastics) are variably silicified. These bodies occur as flat lenses and vein-like vertical-subvertical bodies. They are surrounded by a quartz stockwork. The ore is confined to the silicified and argillic zones and formed in three stages (galena+sphalerite with some Pb, Se and Ag, Sb sulphosalts – tetrahedrite – pyrite with rare Au, Ag, Bi, Te and Sb minerals, e. g. stibnite). See MOLNÁR et al. (1999) for details and further references.

A paragenesis rich in secondary minerals has been found recently on the southern slope of the Fehér-kő Hill, in the cavities of intensively leached siliceous-baritic vein fillings above the Egyesség gallery. Antimony oxides present are stibiconite and bindheimite according to the XRPD and EDS studies. Characteristic reflections of stibiconite are 5.96 (55), 3.11 (+barite), 2.978 (100), 2.581 (20), 1.820 (+quartz) and 1.545 (+quartz) [d(Å) (I_{rel})]. Stibiconite forms yellowish white to light brown, firable, porous coatings on the cavities of the quartz veins. Bindheimite appears in yellow, porous, earthy coating. Based on XRPD and EDS analyses, the secondary minerals of the paragenesis are as follows: jarosite, scorodite, mimetite, malachite, cornubite, richelsdorfite, olivenite, azurite, gypsum, goethite, beudantite/segnitite and smithsonite (?). Antimony oxides and arsenates have been formed by weathering of tetrahedrite and tennantite.

NAGY (1985) reported the occurrence of stibnite grains few mm in size on the rim of pyrite and as inclusions in pyrite-bearing samples collected on the waste dump of the Hegyes Hill Gallery and studied by microprobe.

a) Gyöngyösoroszi, polymetallic lead-zine deposit

The Gyöngyösoroszi–Mátraszentimre deposit crops out over a 12 km^2 area between the two villages. Some 20 hydrohtermal veins are hosted by andesitic lava flows and pyroclastic rocks of the Middle Andesite Sequence. The predominant trends of the veins are NNW–SSE, NNE–SSW and N–S. The (siliceous-carbonate) veins show symmetrical infillings with a variety of textures. The veins developed in seven stages with pyrite, marcasite, sphalerite, wurtzite, galena, and chalcopyrite as main products in the most important sulphide-bearing periods (2–4 stages). See MOLNÁR et al., 1999 for details and further references.

In Hungarian stibnite is the most frequent and the most varied in appearance in Gyöngyösoroszi. The oxidation zone of the deposit is shallow; therefore secondary antimony oxides appear only in minor amounts. Cervantite (NAGY, 1986) and valentinite (KOCH, 1966) have been reported so far.

Before describing the antimony oxides of the deposit, we give a list of the workings where stibnite was found according to KUN (1985). Stibnite is known from vein fissures at the +465 m level of the Károly vein as acicular aggregates on quartz or inclusions in calcite crystals. In the northern part of the Károly vein, above the +430 m level, stibnite was found in several places as mm- or cm-sized globular aggregates, which were usually embedded in calcite or in clay. The radial-fibrous structure of these dull aggregates could be observed on the fractures. Stibnite in vein fissures of the +470 m level of the Károly vein was accompanied by cinnabar.

Stibnite occurrences of the Új-Károly vein were very similar to those of the Károly vein. In the southern and central sections of the Northern Aranybánya-bérc vein stibnite-rich vugs of amethyst and calcite were found in the cementation zone above the +460 m level. Finally, in the Hidegkút vein stibnite was found associated with barite in the exploration adit and in the workings above the +400 m level.

According to KÁROLY NAGY (personal communication, 1996) stibnite was frequently found in the upper levels of the mine, in the adit until Mátraszentimre, in the Bányabérc, Kiskút and Hidegkút veins and in the compressor hall grown on or within quartz (amethyst). On the +350 m level at about 300 from the shaft a cavity was found during the abandonment of the workings, with an egg-sized, plumose mass of stibnite. In 1980, a dip heading was driven from the +150 m level and a mine working was introduced on the +100 m level for the Arany-Péter vein. Here some cavities contained plumose stibnite aggregates 3–5 cm in diameter (studied by DÓDONY, 1986) on white or colourless calcite. Calcite was sometimes nearly black due to the stibnite inclusions (also reported by KUN, 1985). Stibnite has also been observed in the exploration adit above the Károly gallery on the +560 m level. The mineral was enclosed in quartz and covered by limonite.

According to KUN (1985) rhe greatest amount of antimony ochre was found around the +510 m level in the rise driven to the surface along the Károly vein. Another parageneses that have been observed on the samples of the HOM collection include:

Radial-fibrous stibnite spherules 2–5 mm in diameter with a pyrite-marcasite coating, the latter altered to different sulphates (rozenite, gypsum, and jarosite), on a quartz crust.

Radial-fibrous stibnite aggregates with siderite spherules ("spherosiderite") 0.5–2 mm in diameter, in quartz veins. Sometimes siderite forms coatings on the stibnite aggregate, similarly to the classical stibnite–spherosiderite paragenesis of Baia Sprie (Felsőbánya).

It can be concluded that stibnite occurs frequently in the upper horizons of the Gyöngyösoroszi ore deposit and this paragenesis is related to the final stages of the ore formation (VIDACS, 1961a, GATTER, 1986; MOLNÁR et al., 1999). Associated minerals are calcite, quartz, barite, marcasite/pyrite, siderite, cinnabar; secondary minerals include sulphur, rozenite, gypsum, jarosite, stibiconite and rarely cervantite (see below).

Analytical results of antimony oxides from the ore deposit of Gyöngyösoroszi can be summarised as follows. Usually diffuse peaks of poorly crystallised phases appear on the records; however, stibiconite can be proved in several cases. HOM 18671 specimen deserves special attention. The cream-coloured, thin crust on stibnite is composed of stibiconite with some cervantite. All major reflections of stibiconite appear on the XRPD record (sample D15, Table III), which makes its identification certain. A few small peaks corresponding to medium-intensity reflections of cervantite (Table III) suggest that cervantite may also present in the sample, although the two highest peaks of cervantite are overlapped by two strong reflections of stibiconite. This is the first occurrence of cervantite in Hungary that is supported by XRPD data. It is to be noted that we were unable to detect valentinite although KOCH (1966) had reported it from the Károly vein.

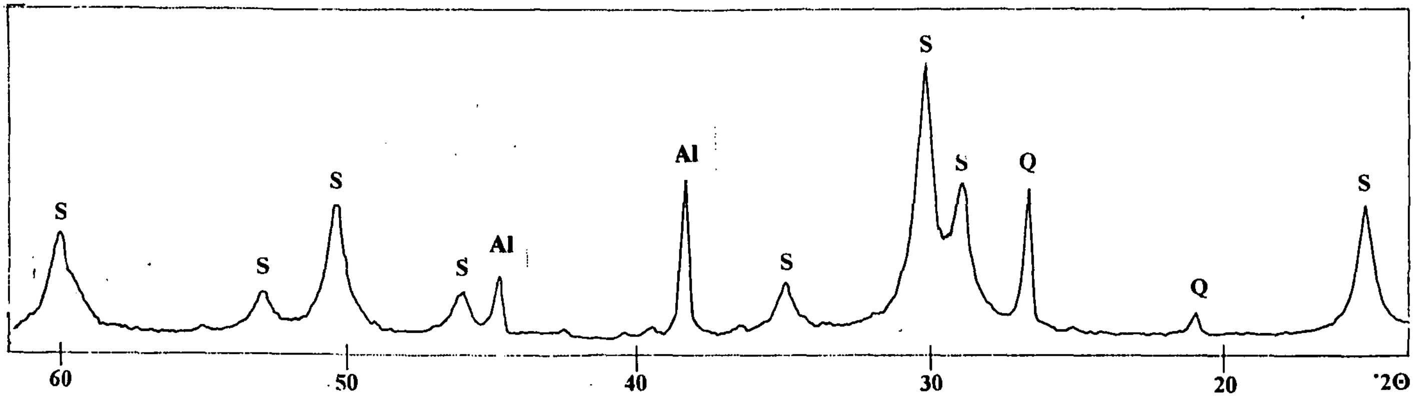

Fig. 4. XRPD pattern of stibiconite from Teréz Hill, Mátraszentimre (C3)
Symbols: Al: sample holder, Q: quartz, S: stibiconite

TABLE 3

XRPD data of an antimony ochre sample from Gyöngyösoroszi (D15)

HOM 18 671 (sample D15)		stibiconite JCPDS 10-388		cervantite JCPDS 11-694		Quartz JCPDS 5-490	
d (Å)	I/I_{max}	*d* (Å)	I	*d* (Å)	I	*d* (Å)	I
5.922	45	5.93	90				
4.254	15					4.260	34
3.567	2						
3.470	8			3.443	35		
3.346	70					3.342	100
3.225	6						
3.097	57	3.09	70	3.073	100		
2.966	100	2.96	100	2.942	45		
2.666	3			2.650	25		
2.569	19	2.57	40				
2.461	7					2.458	12
2.413	1			2.404	17		
2.357	3						
2.284	4					2.282	12
2.241	3			2.235	11	2.237	6
2.130	5					2.127	9
2.097	1						
1.977	15	1.98	30			1.979	6
1.869	2			1.862	25		
1.816	4	1.81	80	1.781	20	1.816	17
1.736	12	1.74	30	1.723	20		
1.707	3			1.697	11		
1.673	2					1.672	7
				1.636	11		
1.566	6	1.57	20				
1.549	27	1.55	60			1.541	15
1.482	6	1.48	30	1.487	13		
1.456	2			1.469	11	1.453	3

(Registered in the X-ray Laboratory of MOL Rt. Budapest, by É. MARGITICS-SIPŐTZ. Remarks: Reflections exceeding 10% relative intensity only are listed from the cervantite JCPDS pattern. Observed d values are corrested using quartz as internal standard.)

b) Mátraszentimre, polymetallic lead zinc deposit

According to NAGY and BARBÁCSI (1966) stibnite occurs here in radial fibrous aggregates and acicular crystals in the vein quartz, associated with pyrite and less sphalerite and galena. In the near-surface samples stibnite is replaced by antimony ochres along the cleavage planes. KOCH (1966) regarded the oxidation product of stibnite as cervantite, which may occur as pseudomorphs as well. This identification has not been confirmed by the present study (see below).

According to KUN (1985) fine acicular stibnite aggregates were found in the southeastern section of the +467 m and +515 m levels of the Mátraszentimre vein, filling fissures that cross the vein. KÁROLY NAGY (personal communication, 1996) reported stibnite from the +864 m level and from the ventillation shaft.

Based on museum samples (HOM) the largest (3-4 cm long) documented individual stibnite crystals of Hungary, although without terminal faces, were found at Mátraszentimre. Secondary antimony oxides in various shades from white to yellow frequently occur in the specimens. A yellowish crust on a sample from the MÁFI collection (inv. # 11677) from the left drift proved to be stibiconite according to the XRPD pattern (sample # C3, Fig. 4).

On the southern slope of the Teréz Hill exploration trenches were driven by Aladár Vidacs in the 1950s (VIDACS, 1961b). On the basis of the (MÁFI) museum samples stibnite was found abundantly in these trenches. The mineral usually formed typical radial-fibrous aggregates in the quartz veins of the altered andesite. One sample of this vein-filling (MÁFI, inv. # 2946) contains a 7–9 cm thick aggregate of stibnite and white and butter yellow antimony oxides. Another specimen contains stibnite pockets 1–3 cm in diameter, rimmed by white to light yellow antimony oxide coatings 1–2 cm thick, with pseudomorphs 2-5 mm long, after stibnite in the cavities. The secondary antimony oxide of these specimens proved to be stibiconite on the basis of the XRPD patterns (samples D10 and D12). Characteristic reflections are 5.88 (80), 3.084 (70), 2.956 (100), 1.809 (+quartz), and 5.92 (55), 3.09 (70), 2.963 (100), 2.568 (20), 1.820 (40), 1.546 (+quartz) [d(Å) (I_{rel})], respectively. Reflections of stibnite and/or quartz have also appeared on the patterns.

Asztag-kő Hill, Gyöngyössolymos, antimony mineralisation in silicified rhyolite tuff

In Hungary antimony oxides have been found the most abundantly on the Asztag-kő Hill at Gyöngyössolymos in the upper horizon of the hydrothermally silicified rocks related to a steam-heated alteratation zone of a low-sulphidation type epithermal system. The siliceous rock is dark grey due to embedded stibnite on the western side of the steeply dipping kaolin vein near the benchmark on the top of the hill. Stibnite crystals up to 5 cm long used to be found near the surface (SZUROVY, 1940). KISS (1960) noted that stibnite is usually coated "by yellow antimony-ochre, cervantite". KOCH (1966) added that the cervantite pseudomorphs in small vugs are dark straw-coloured, whereas those after stibnite embedded in the rock are light yellow. Koch also reported the rare occurrences of sénarmontite. Small octahedral crystals with diamond lustre were found associated with cervantite pseudomorphs in small vugs. In a brief report on the paragenesis SZAKÁLL (1989) described stibiconite on the basis of XRPD data, and postulated the presence of tripuhyite indicated by EDS analyses.

The appearances of the antimony oxides was highly variable in the several hundred samples examined by optical microscope. The colours ranged from white thorugh butter yellow, lemon yellow, orange and light brown shades to reddish brown and dark brown, sometimes even to black. EDS analyses revealed a definite relationship between iron content and colour; viz. darkes shades correspont to higher iron content (cf. VITALIANO and MASON, 1952). The hardness of the antimony oxides varies between 2 and 4 Mohs' scale. The morphology is also varied: thin to thick crusts with smooth to rough surfaces (*Fig. 5.*), globular, stalactitic or irregular aggregates. Every intermediaries from fresh stibnite to crystals that completely altered to antimony oxides (stibiconite, tripuhyite) including common pseudomorphs after stibnite (Fig. 6) can be observed. From among antimony oxides only sénarmontite forms well-developed crystals; colourless or white octahedral of 1–3 mm size and crusts composed of octahedral of 0.05–0.1 mm size also occur (*Fig. 7*). A pale purple aggregate of 0.1 mm-long acicular crystals was also observed once. The habit and the qualitative EDS data (Sb+, S+; O cannot be detedted due to the experimental conditions) suggest kermesite but in lack of enough material it was impossible to confirm by XRPD.

About half of the examined samples were difficult to identify by XRPD, as they are poorly crystalline, their reflections are diffuse. In other cases sharp peaks of stibiconite can be observed (e.g. sample V204). Well and poorly crystalline phases proved to be indistinguishable by their appearance. Cervantite and valentinite have not been detected undoubtedly in any sample.

As far as the presence of tripuhyiite is concerned, it has already been suggested on the basis of the high iron content of some reddish brown and dark brown antimony oxide phases (SZAKÁLL, 1989). One of the XRPD patterns made during the recent study (sample D37) showed broad reflections of tripuhyite accompanied by those of quartz. On other patterns a few, ambiguous, low-intensity peaks of tripuhyyite appeared together with strong reflections of stibiconite.

Antimony oxides of the antimony-bearing paragenesis of the Asztag-kő Hill are associated with quartz, barite, calcite, pyrite/marcasite, sphalerite, cinnabar, galena, anatase, goethite, hematite, manganese oxides, pharmacosiderite, scorodite (?) and sulphur.

Parádsasvár, stibnite occurrence in rhyoite tuff

NAGY and SZENTES (1969) found thin quartz veins with fine needles of stibnite about 500 m east from the village, in the cut of the Parád-Gyöngyös road. The surface of stibnite was reported to have been covered by cervantite according to the XRPD data (unpublished). On the original sample (MÁFI 10399), 2–4 mm long radial-fibrous stibnite aggregates appear in thin quartz veinlets in intensively weathered rhyolite tuff, which contains lemon yellow spots of jarosite. A film-like, greyish-white weathering crust rims the stibnite crystals. According to the XRPD investigations (sample D13), the weathering product is stibiconite rather than cervantite. Characteristic reflections are 5.92 (60), 3.06 (+stibnite), 2.962 (100), 2.564 (80), 1.811 (+quartz) [d(Å) (I_{rel})].

Gyöngyös-Mátraháza, stibnite occurrence in andesite

The HOM collection holds a stibnite sample (inv. # 22661) collected by Péter Badinszky, its locality is given as "Mátraháza, road cut". This occurrence doesn't correspond to the above-described one, since the host rock of the stibnite-bearing quartz veins is altered, pyretic andesite and not rhyolite tuff. In some parts of the quartz veins, columnar stibnite crystals are embedded in calcite. Calcite is black due to another fine acicular stibnite generation. A brown film of antimony ochre covers the surface of some stibnite crystals. The weathering product cannot be identified by XRPD due to its poor crystallinity.

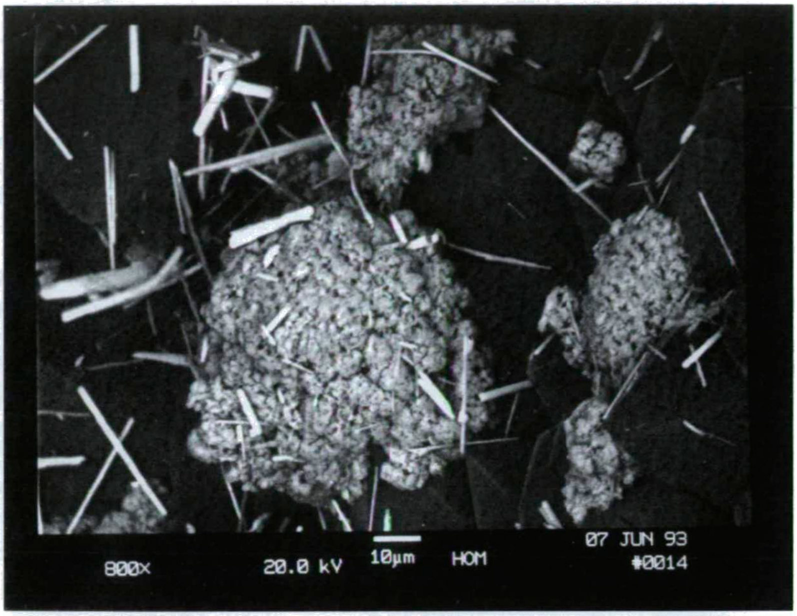

Fig. 5. Tripuhyite aggregates (grey) with stibnite needles (white) on quartz (dark grey) from Asztag-kő Hill, Gyöngyössolymos, SEM image

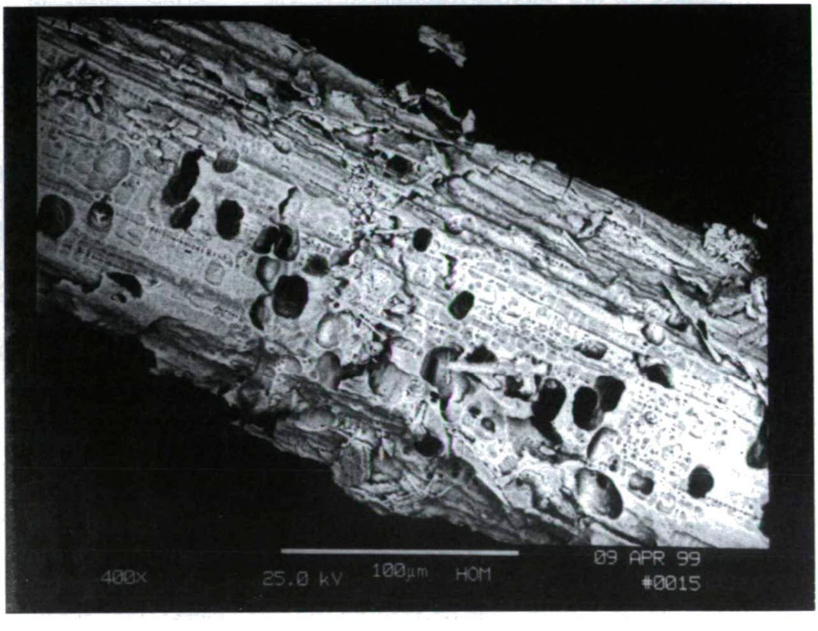

Fig. 6. Stibiconite, pseudomorphs after stibnite, Asztag-kő Hill, Gyöngyössolymos. SEM image

Fig. 7. Sénarmonite crystals, Asztag-kő Hill, Gyöngyössolymos. SEM image

Rudabánya Mts.

The Rudabánya Mts. forms the Eastern part of the Aggtelek–Rudabánya Mts., which is one of the most complex regions of Hungary considering its geology and structural development. The Rudabánya Mts. Is built up by four dislocated segments and itself is separated from the Aggtelek Mts. By a lateral dislocation zone. In the basement an Uppermost Permian and Lowermost Triassic evaporitic sequence is found, which is followed by Lower Triassic to Jurassic shallow to deep marine formations (marl, limestone, dolomite, etc.). The cover formations are Miocene to Pliocene sediments. There are several metasomatic iron ore occurrences in the Middle Triassic formations of the mountain.

Rudabánya, iron ore deposit

The large siderite body at Rudabánya, which is exposed in some 4 km length by the huge open pits was formed by the metasomatism of a Triassic sedimentary series built up by Lower Triassic marl and limestone and Middle Triassic (Gutenstein) dolomite. Subsequent hydrothermal events produced a multiphase sulphide mineral paragenesis. The amount of sulphides can be estimated from the historical fact that Rudabánya used to be a rich silver and copper mine in the Middle Ages.

The ore-forming phases and their products are reconstructed as follows (see PANTÓ, 1956; JANTSKY, 1966; HERNYÁK, 1977 for further details and references). In the first phase a hematitic and slightly sideritic ore formed with some chalcopyrite along the main overthrust planes. During the second (main iron producing) phase tectonised dolomite and limestone turned into a so-called sparry iron ore (siderite, ferroan dolomite, tha latter also called incorrectly "ankerite"). In the third phase vein- and stockwork-like bodies were formed. A peculiar product is a 0.5 to 3 m thick barite zone ("marginal barite") with abundant sulphides, which was formed a on the margin of the marl. The ore formation is terminated by later large-scale weathering processes producing ochreous, siliceous and brown iron ore. A peculiar secondary product is the so-called spherosiderite ore (from the radial–spherical texture of siderite grains under the microscope), which was formed by the reprecipitation of dissolved iron.

As a result of analyses and field observations made in the last decade, minerals such as stibnite, stibiconite, valentinite, bindheimite and a red, unidentified antimony oxide phase were detected from several outcrops of the deposit.

Stibnite was found in two different parageneses in Rudabánya. It appears in radial aggregates of 1–3 mm long crystals in the outcrops of the spherosiderite ore (Polyánka area and Andrássy III mines) together with siderite, calcite, malachite and realgar. (It is to be noted that the miner's term to the spherosiderite ore is "scoriaceous ore" because of its porous nature, see PANTÓ, 1956.) This stibnite-bearing paragenesis belongs to the last sulphide phase of the ore formation. Secondary antimony oxides have not been found in this paragenesis. Stibnite of the second paragensis forms sheaf-like aggregates of 1–2 mm long crystals in the galena- and sphalerite-bearing ores of the large baritic zones in the Polyánka area. Here stibnite is covered by white crusts composed of valentinite (see below).

Bindheimite occurrences are of two types. Bindheimite is ubiquitous in the weathered galena-sphalerite ore of the large barite bodies. Lemon yellow to orange, powdery crusts covering up to 10–20 cm^2 areas, as well as earthy, irregular aggregates of 1–2 mm size were discovered in the Polyánka, Andrássy I, Andrássy II, Villanytető and Vilmos mining areas. It was also found rarely as yellow, acicular crystals and radial-fibrous aggregates, which may be pseudomorphs after heteromorphite, which was also observed here. Associated minerals (Villanytető area) are cerussite and barite according to and XRPD record (sample L55). The second type of occurrence is in the silicified limonite of the Adolf area. Here bindheimite forms yellow, earthy crusts in the fissures of the ore. Parent sulphide minerals have not been found even in the wider environment of bindheimite. It is to be noted that KERTAI (1935) mentioned native sulphur crusts 0,5–1 mm thick usually covered with a cinnabar film associated with cerussite. We think that this "sulphur" occurrence may have been bindheimite in fact.

Stibiconite is also widespread, although it occurs in much smaller amounts. It forms radial-fibrous aggregates of white colour and diamond lustre after stibnite in fissures of quartz veins in the siliceous limonite of the Adolf area. Every important reflection of stibiconite appeared on the XRPD pattern (sample G163) 2.978 (100), 1.822 (+quartz), 3.11 (35), 5.95 (30), 2.578 (30), 1.554 (30) etc. [d(Å) (I_{rel})]. It is also connected to the fissures of intensively silicified zones of limonite at the upper horizons of the Villanytető area, where stibiconite forms white, 0.5 to 1 mm thick crusts and globular aggregates. Acicular pseudomorphs after stibnite are found in the cavities of the spherosiderite ore of the Andrássy III mine. The most abundant accompanying minerals in these outcrops are malachite, azurite, goethite and cinnabar. In a sample from the Villanytető area white spherules 0.2–0.4 mm in diameter of an antimony oxide (by EDS) are found on azurite crystals associated with calcite in cavities of limonite.

Partzite forms olive green powdery aggregates or friable coatings in the cavities of the so-called spherosiderite ore of the Andrássy III mine. SEM micrographs revealed that the coatings are composed of isometric graisn up to 1-2 μm without any apparent crystal forms (*Fig. 8*). Only the strongest reflection of partzite is observalble on the XRPD pattern (sample K281) as a broad peak at 2.96 Å. Cu and Sb content was proved by EDS. Associated minerals are malachite, cuprite, native copper, siderite and quartz in the samples. Its formation may be related to the weathering of chalcostibite and other Cu, Sb sulphides that are present in the spherosiderite ore.

Valentinite is known only from the Polyánka area as a rarity. It forms crusts of white euhedral crystals with diamond lustre on acicular stibnite aggregates. The crusts are composed of 0.1–0.3 mm long bipyramidal crystals according to the SEM observations (*Fig. 9*). The identification was confirmed by XRPD (*Fig. 10*). It is to be mentioned that this is the first report of valentinite from Hungary based on XRPD and EDS results. Further members of the secondary mineral paragenesis that appears in the weathering zone of the Polyánka galena-sphalerite-bearing barite body are cerussite, gypsum, mimetite, jarosite, goethite, acanthite, barite, bindheimite and smithsonite.

Based on EDS analysis, an antimony- and sulphur-bearing secondary mineral appears also in a secondary baritic fissure filling in the Polyánka area, forming red, earthy crusts and, very rarely, irregular aggregates up to 0.1 mm size. SEM images shows that the aggregates consist of foraminous spherules 5-10 μm in diameter (*Fig. 11*). An X-ray study was attempted using Gandolfi camera without interpretable results.

Martonyi, iron ore deposit, doubtful stibnite occurrence

A small iron ore deposit, similar to that at Rudabánya is found near to Martonyi. Middle Triassic Gutenstein dolomite was replaced metasomatically by the main "primary" minerals including siderite, quartz, barite, pyrite, chalcopyrite, bornite, tetrahedrite and galena; secondary products are goethite, hematite, chalcocite, covellite, malachite, azurite and cerussite, etc. (JANTSKY, 1966).

Maderspach (1880) listed stibnite among the minerals of Martonyi. Although stibnite really occur in the above-described Rudabánya deposit of similar origin, the size of the Rudabánya stibnite suggests that Maderspach probably confused manganese oxides with stibnite as it had happened earlier in the case of the Úrkút pyrolusite (cf. PAPP, 1990).

Tokaj Mts.

The main mass of the mountain is made up by Badenian to Lower Pannonian andesite-rhyolite volcanic rocks related to horst/graben structures with interbedded remnants of small andesite stratovolcanoes, dacite extrusive domes and extensive rhyolite pumice tuff horizons with flow-dome complexes in volcanic centres. There are geophysical evidences of diroite porphyry intrusions. Volcanic rocks rest upon Middle Miocene sedimentary rocks, Hercynian to Late Palaeozoic basement is exposed on horsts. Ore deposits and showings are bound to intrusions-related Au-Ag±Sb low-sulphidation epithermal veins and stockworks and steam-heated alteration zones. See MOLNÁR et al. (1999) for details and further references.

Erdőbénye, antimony showings in lacustrine siliceous sediments (limnoquartzite)

Between Erdőbénye and Sima a limnic sequence, composed from rhyolite tuff, tuffite, sand, sandstone, diatomite and quartzite, is found. The limnic sequence is exposed by boreholes and diatomite open pits. The quartzite used to have been quarried for millstones. (See MÁTYÁS, 1979, for further details.)

SZABÓ (1870) described a stibnite vein in limnoquartzite from a millstone quarry west from Erdőbénye in the valley of the Sás Stream. PAPP (1982) investigated a museum specimen (ELTE) in detail. He observed thin, light brown crusts of antimony ochre

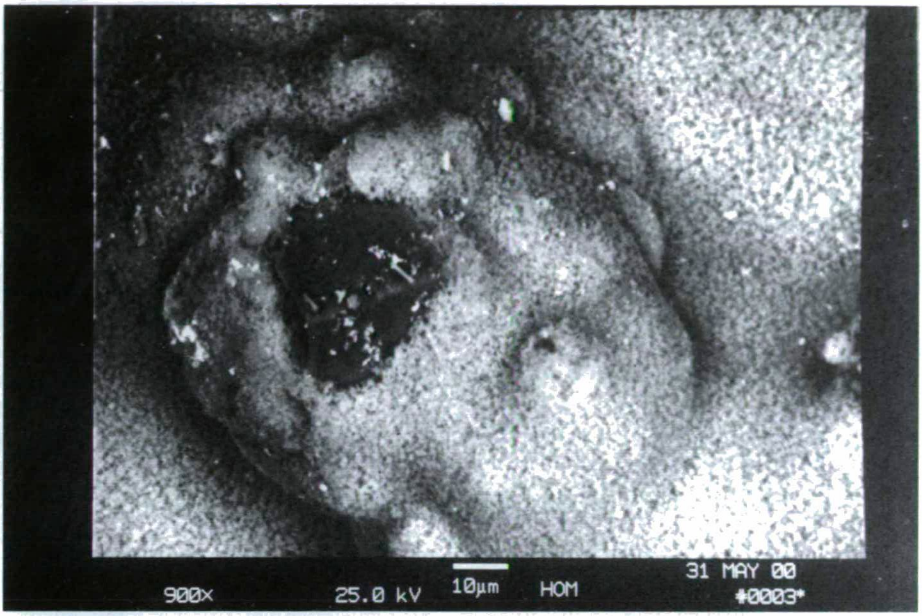

Fig. 8. Partzite crust on siderite rhombohedra, Andrássy III mine, Rudabánya. SEM image

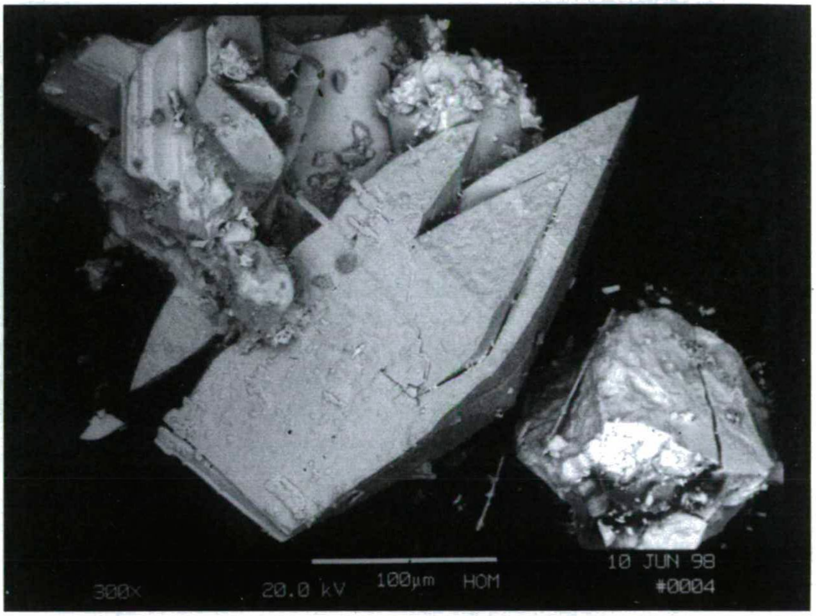

Fig. 9. Valentinite, bypiramidal crystals, Polyánka area, Rudabánya, SEM image

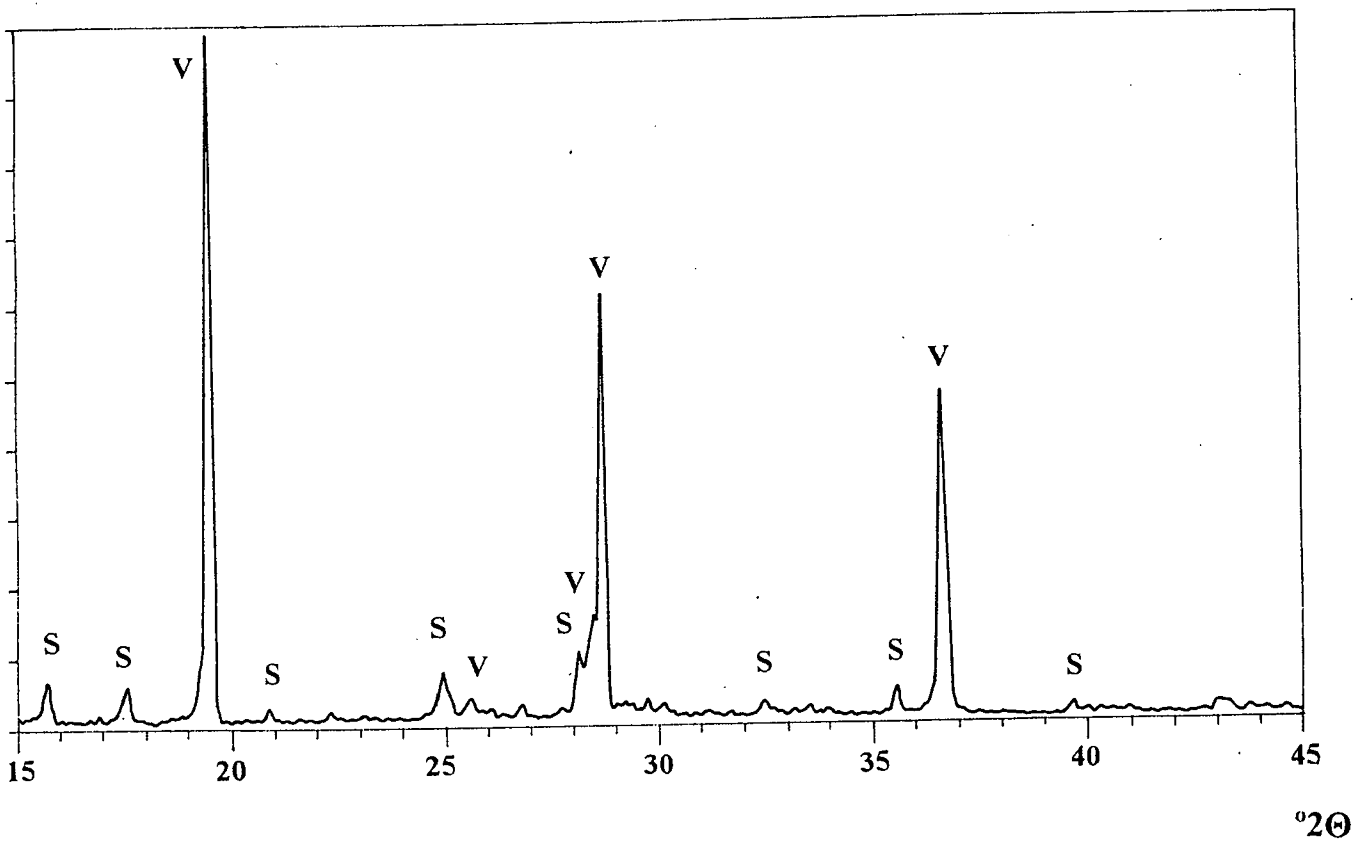

Fig. 10. XRPD pattern of valentinite, Polyánka area, Rudabánya. Symbols: S: stibnite, V: valentinite

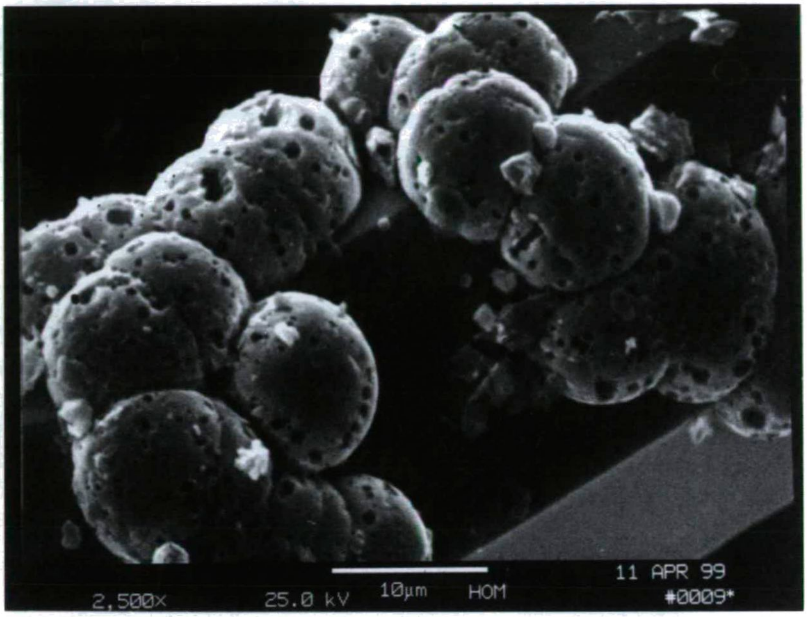

Fig. 11. Foraminous aggregate of an unidentified secondary antimony mineral, Polyánka area, Rudabánya

rimming the stibnite vein, which was embedded in silica minerals, quartz (chalcedony) and opal. In polished sections, the antimony ochre appeared as pseudomorphs after stibnite, forming white, radial aggregates of acicular crystals up to 1 mm long. The mineral is probably stibiconite on the basis of the XRPD patterns. Framboidal pyrite aggregates were also observed as two concentric strings along the inner part of the stibnite vein.

ENDES (1988) found a similar occurrence in the debris of a temporary steam on the western slope of the Mogyorósok Hill; most of the stibnite enclosed in blocks of limnoquartzite is exposed in the bed of a temporary stream between the Liget and Mogyorósok areas. Stibnite veins 1-2 cm in thickness are found on the bedding planes of the thin-bedded limnoquartzite. Peripheral parts of the veins are usually covered by yellowish white to light brown antimony oxides. In more disturbed zones, where weathering proceeded along the joints, the alteration of stibnite is more pronounced. Stibiconite was identified by XRPD; weak but characteristic reflections were observed in addition to those of stibnite (sample G2). Sb, Pb, and Cu were detected on the XRF spectrum. In another sample (EBSZ-1S) the broad reflections of stibiconite (5.91 (50), 3.09 (60), 2.95 (100), 2.56 (25), 1.81 (30) [d(Å) (I_{rel})]) were accompanied by quartz and opal peaks. The samples contained pyrite framboids similarly to those found in the sample from the Sás Stream valley. All stibnite occurrences associated with limnoquartzite near Erdőbénye are supposed to have been formed by epithermal processes. Later stibnite altered more or less to stibiconite at near-surface conditions. It should be mentioned that according to ERNŐ MÁTYÁS (personal comm.) small amounts of stibnite were found several times in limnoquartzite beds traversed by the prospect boreholes of the Ligetmajor diatomite deposit.

Herczegköves and Koldu open pits, Rátka, antimony showings in lacustrine siliceous sediments (limnoquartzite)

Between Mád and Rátka a sedimentary sequence of a lacustrine basin crops out over an approx. 8 km^2 area (see MOLNÁR et al., 1999 for details and further references). The limnic sequence of the basin is underlain by a pyroxene andesite lava flow. In the lacustrine sequence 3 major consolidated siliceous layer occur. The siliceous layers show bedding from few cms up to 1–2 m. Bentonite, bentonitic tuffite, rhyolite tuff etc. are interbedded. Siliceous beds show high concentration of Sb and less As and Hg, as well as some Ag (VETŐ, 1971).

A stibnite occurrence, similar to those around Erdőbénye, is known from the limnoquartzite cover of the bentonite open pit of the Hercegköves area (misnamed for Koldu by JÁNOS and PAPP, 1985). Radiating circular or spherical aggregates up to 3 and 1-cm diameter, respectively, are found on the bedding planes of the rock and in the irregular cavities within the beds. Stibnite clusters have usually been altered to whitish to ochre or brownish yellow stibiconite (investigated by XRD and EDS by JÁNOSI and PAPP, 1985).

MASON and VITALIANO (1953) suggested that "the brown or black colour of some stibiconites is generally due to the presence of admixed tripuhyite". Recent XRF studies showed that the darker stibiconite samples from Hercegköves have a higher iron content than that of the lighter ones. The XRPD pattern of the darker samples corresponded to that of stibiconite but the increase of width and relative intensity of the peaks at 2.56 and 1.72 Å suggested the presence of a poorly crystalline tripuhyite-like phase (the strongest tripuhyite reflection at 3.28 Å was overlapped by the large quartz peak at 3.33 Å). A brown, friable, earthy substance filling a cavity showed the highest iron content. The sample gave an XRPD patterns with broad reflections of tripuhyite, some small peaks of stibiconite, opal and quartz, and probably some poorly crystalline goethite (*Fig. 12a*). After heating to 1000 °C tripuhyite peaks became sharp and well defined, the crystallinity of opal also increased (*Fig. 12b*). The most intensive reflections of hematite appeared on the pattern, whereas no traces of stibiconite were visible. In a parallel experiment an iron-free sample of stibiconite + quartz gave identical XRPD patterns before and after heating.

It is to be noted that pyrite hexahedra, cinnabar and realgar have also been found in the close environment of the stibiconite.

There are similar specimens in the MÁFI collection (collected by ORSOLYA KÁKAY SZABÓ in 1971) from the near Koldu open pit, suggesting an occurrence identical to that at Hercegköves.

Telkibánya, Au-Ag ore deposit

The most renowned ore deposit of the Tokaj Mts. is that at Telkibánya. The mining flourished in the Middle Ages with a short-term revival in the 18th century. Badenian volcanic rocks host small veinlets of the older stockwork-type mineralisation (sphaerlite, galena, pyrite, chalcopyrite, and arsenopyrite). A younger and more important near-surface mineralisation with abundant pyrite dissemination in the hydrothermally altered Sarmatian volcanic rocks and noble metals in siliceous veins consists the major part of the low-sulphidation type epithermal deposit.

"Antimony ochre" was mentioned from Telkibánya by ZIPSER (1817) and by COTTA and FELLENBERG (1862). This piece of information has been republished in several monographs in a rather confusing way. ZEPHAROVICH (1859) at first simply adopted Zipser's data on antimony ochre but later (ZEPHAROVICH, 1873) he reclassified it as cervantite without any any further investigation, referring to the studies of Blum on

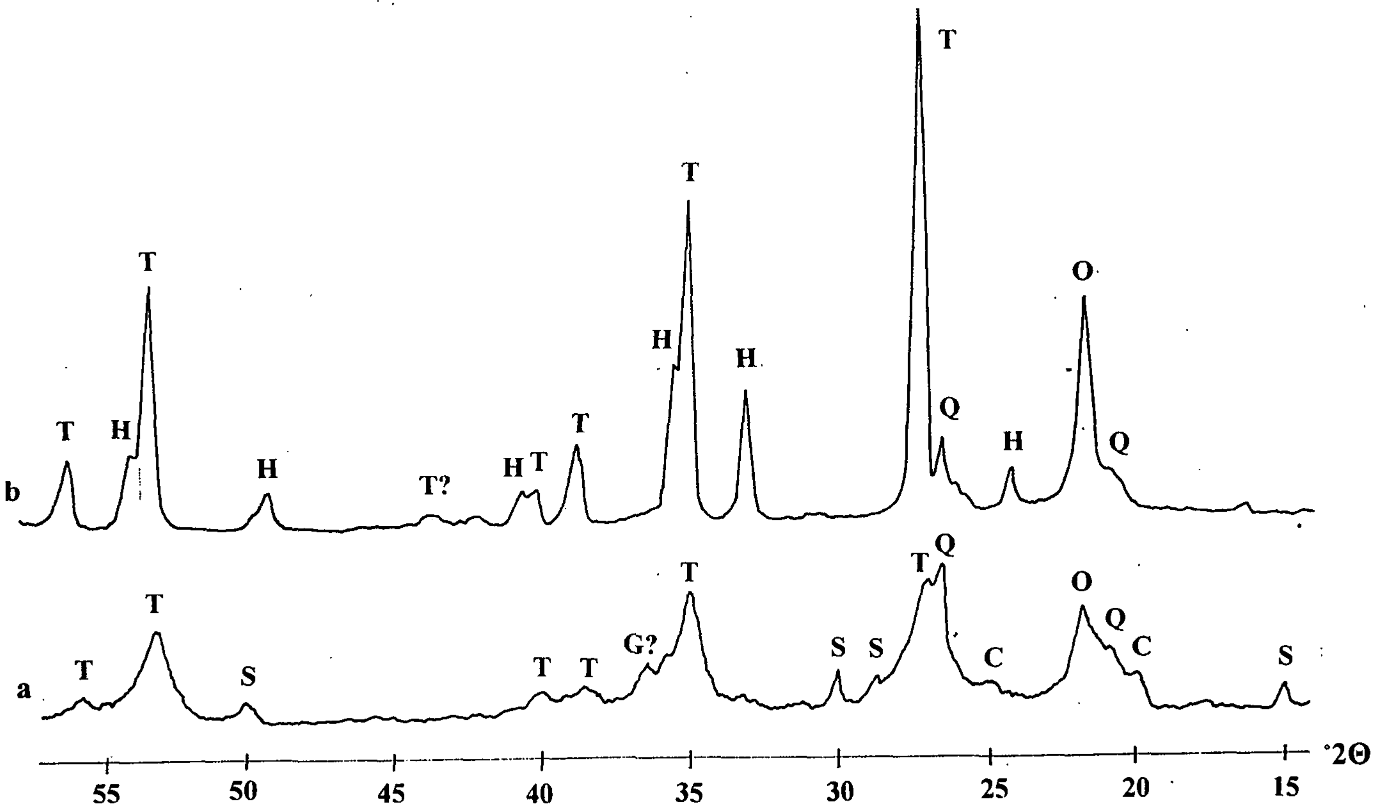

Fig. 12. XRPD pattern of tripuhyite, Hercegköves open pit, Rátka (a) before and (b) after heating to 1000 °C. Abbrevations: C: a clay mineral (d001≈7.1 Å), G: goethite, H: hematite, O: opal, Q: quartz, S: stibiconite, T: tripuhyite

antimony ochre from other localities. TÓTH (1882) published the data of Zipser and of COTTA and FELLENBERG (together with those from other Hungarian occurrences) under the heading "antimony ochre", noting that he used this general term for the mineral because it was not known which one is cervantite and which is stiblite [=stibiconite]. LIFFA (1955) gave antimony ochre, cervantite and stiblite alike. In the first edition of this monograph KOCH (1966) pointed out that "one failed to find the antimony ochre mentioned by earlier authors" but, curiously enough, he included cervantite in the paragenesis referring to TÓTH (1882). Antimony ochre has not been found even during the detailed study of the ore deposit by SZÉKY-FUX (1970). Cervantite and antimony ochre were then omitted from the Telkibánya chapter of the second edition of the monograph of KOCH (1985). MOLNÁR and SZAKÁLL (1994) were also unable to find any antimony oxides in the part of Telkibánya mining area, except for a light yellow, unidentified iron- and antimony-bearing oxide from a pit on the Fehér Hill. They suggested that early authors might have been mistaken acanthite for stibnite and jarosite for antimony ochre.

For only documented stibnite specimen form the Telkibánya area was collected from the spoil heap of the Nyíri gallery on the Fehér Hill, west from Nyíri (HOM collection, inv. # 24 404). Fibrous aggregates of 1–3 mm long crystals were found in cavities of vuggy vein quartz. A yellowish antimony oxide coating covers the surface of the crystals. The poorly crystalline material cannot be identified by XRPD.

A forgotten antimony occurrence from the Tokaj Mountains (?)

Some mineral collections (HOM, Pannonhalma Abbey and a former old private collection) hold similar stibnite specimens collected presumably in the 18–19th century; strangely enough, neither of them has exact locality data. On an old label attached to one of the specimens (HOM, formerly Pál Kriván's collection), the word "Tokaj" may be rad (with a later question mark pencilled after the name). Another autograph label by Kriván reads: "perhaps Tokaj Mountains?". The outward appearance and the paragenesis of the host rock suggest that the locality was somewhere in the Eperjes–Tokaj Mountains (Slanské Mts. in Slovakia and Tokaj Mts. in Hungary).

Main features of these samples are as follows: stout, columnar stibnite crystals, which as a rule form radial aggregates, are embedded in white to light brown common opal. The crystals are well formed; even their terminal faces are frequently developed. (It is to be mentioned that all other stibnite specimens from the Eperjes–Tokaj Mountains, at least those known by us, consist of fine acicular aggregates.) A white, thin film of antimony oxide may cover the surface of the stibnite crystals; due to its low crystallinity, XRPD results were not interpretable. A yellow powdery material obtained from one of the specimens proved to contain poorly crystalline sphalerite.

Supposing that these samples were really collected from an obsolete occurrence of the Tokaj Mountains, the only locality (to our knowledge) where a considerable amount of stibnite is mentioned from is the antimony mine at the former Klauzúra or Szigord (now Sigord) E from Kokošovce near Prešov (cf. MADERSPACH, 1875; FOULLON, 1884).

SUMMARY AND CONCLUSIONS

General conclusions

Stibiconite proved to tbe ubiquitous among antimony ochre minerals of Hungary. On the other hand, cervantite, which is frequently mentioned in the literature (usually without any reference to analytical data), is actually rare. VITALIANO and MASON (1952) reached a similar conclusion during their global research on antimony oxides from different localities. It can also be stated that in some cases "cervantite" or "antimony ochre" reported in the Hungarian mineralogical papers is in fact jarosite. This misinterpretation may be due to the similarity of appearance and paragenesis of jarosite and of antimony oxides.

Bindheimite is the second most widespread antimony-bearing alteration product. It is usually formed by the weathering of fahlore or galena containing antimony (as sulphosalt or other inclusions). Bindheimite was frequently associated with cerussite and stibiconite.

Valentinite was unambiguously detected only in one (stibiconite-free) sample from Hungary but, as stibiconite strongly prevails in the examined samples, the presence of small amounts of valentinite in other occurrences cannot be excluded (cf. VITALIANO and MASON, 1952).

Sénarmontite and partzite occur as a rarity.

Tripuhyite was found in iron-rich dark parts of stibiconite-dominated associations in two localities.

Antimony oxides rarely form monomineralic aggregates. The weathering product usually contains relics of stibnite, or other admixed minerals, such as quartz, opal, pyrite, marcasite, iron oxides (goethite, hematite), gypsum and jarosite. Sometimes antimony oxides are intermixed, e.g. bindheimitie + stibiconite, stibiconite + cervantite, stibiconite+tripuhyite assemblages have been detected.

The colour of antimony ochres varies from white to dark brown. According to our semi-quantitative electron microprobe (EDS) analyses, the colour of the weathering product practically depends on the iron content: the higher is the iron content, the darker is the antimony ochre (cf. VITALIANO and MASON, 1952).

X-ray powder diffraction (XRPD) analyses indicated that the antimony ochres show different degrees of crystallinity. About one third of the studied samples hasn't produced any interpretable reflections.

Summary of occurrences

Microscopic stibnite grains that occur in the Kőszeg–Vashegy area and in the Sopron Hills are not accompanied by antimony oxides.

In the Velence Hills stibnite is known from two ore deposits (Kőrakás Hill and Szűzvár, both at Pátka) and also from one ore mineralisation (Meleg Hill, Lovasberény). The weathering products of the Kőrakás Hill and Meleg Hill stibnite are stibiconite, presumably as a weathering product of Pb, Sb-bearing sulphides.

In the Börzsöny Mountains stibnite was found as a rarity at Nagybörzsöny (Nb-13 borehole), and a small amount of as yet unidentified antimony oxides was found at Nagybörzsöny ore deposit (Felső Rózsa gallery).

In the Mátra Mountains stibnite is known from the upper horizons of the Gyöngyösoroszi and Mátraszentimre ore deposits, its weathering products such as stibiconite and, in a single case, cervantite were found in some heavily tectonised zones. The stibnite-rich siliceous formations of the Asztag-kő Hill near Gyöngyössolymos hold one of the most varied paragenesis of antimony oxides in Hungary, due to its shallow position. Stibiconite and bindheimite were detected in the rich secondary mineral paragenesis of the outcrops of veins on Fehér-kő Hill at Parádfürdő. Small stibnite occurrences were found in fissures or rhyolite tuff and andesite of the Mátra Mountains and also in the deep-seated parts of the Recsk ore deposit.

In the Rudabánya ore deposit antimony oxides are found in the weathering zone of the sulphide (galena, sphalerite, stibnite, Ag-sulphides) –rich barite bodies (bindheimite, valentinite) or accompany the stibnite of the so-called spherosiderite ore (stibiconite, partzite).

In the Tokaj Mountains near Rátka and Erdőbénye limnoquartzite bodies that were formed during the post-volcanic activity of Miocene volcanism include a characteristic stibnite + framboidal pyrite + stibiconite ± tripuhyite mineral assemblage. Stibnite occurs only in minor amounts in the eastern part of the Telkibánya mineralised are (Nyíri adit); a weakly crystallised antimony oxide phase was found here as a weathering product.

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